LA-UR-23-20396

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Title:	Gas Liberation, Detection, & Quantification from Geological, Experimental, & Nuclear Weapons Test Materials
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Intended for:	Report
Issued:	2023-01-17

Los Alamos NATIONAL LABORATORY

Gas Liberation, Detection, & Quantification From Geological, Experimental, & Nuclear Weapons Test Materials

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01-17-2023

LA-UR-23-######

NNSA National Nuclear Security Administration

LDRD

Part 1

Hayden Miller

Overview of Accomplishments

Task: Establish new capabilities to liberate and quantify gases from test-site materials, including stable isotope measurements on targeted gases

Accomplishments:

- Designed, built, tested, and established crush/gas purification line
- Developed calibration methodology via calibrated volume (Pettit & Schaller, 2019)
- Successfully applied our methodology to analog and experimental materials:
  - Demonstrated gas liberation and quantification from analog geologic (Soda Dam carbonate, trinitite aerobeads) and experimental materials
  - Demonstrated the measurement feasibility of stable isotope analysis of target gas (CO_2) obtained from a crushed material

Refurbished Laboratory Space in the GGRL

New Crushing and Gas Extraction/Purification Capabilities

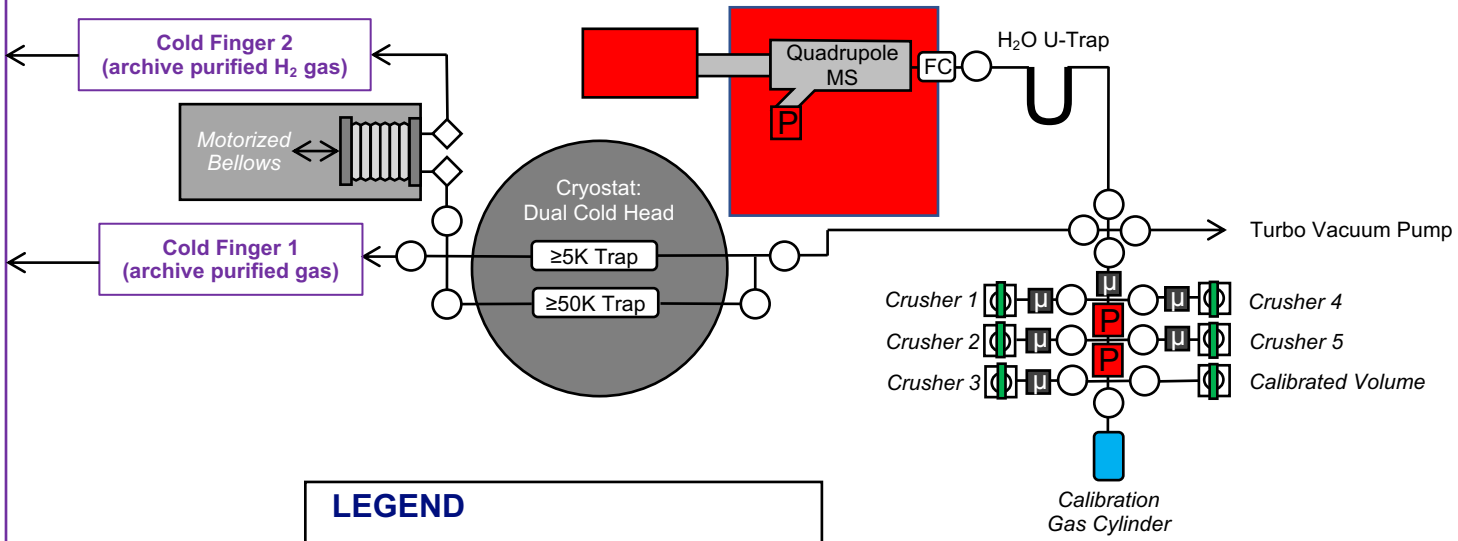
Custom Vacuum Line for Gas Liberation, Quantification, & Purification

Custom Vacuum Line for Gas Liberation, Quantification, & Purification

Isotope Ratio Mass Spectrometry

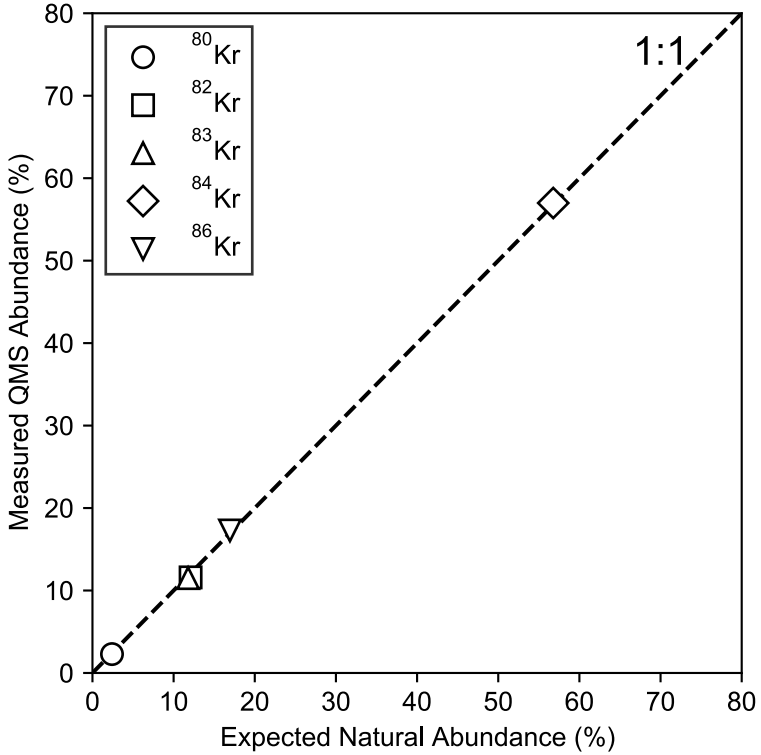
Thermo Delta V Plus IRMS

Thermo MAT 253 IRMS



QMS Noble Gas Quantification: Krypton Stable Isotopes

Sample size: 0.2 to 2 nanomoles (10^{-9}) pure Kr gas

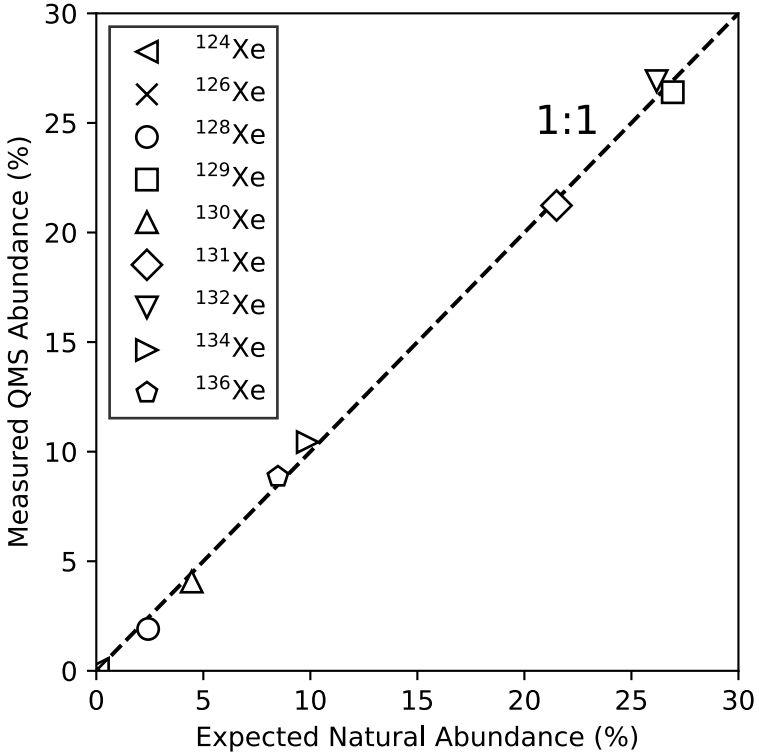


Preliminary External Reproducibility (1σ; *n* = 5)

^{80}Kr: ± 0.13%
^{82}Kr: ± 0.15%
^{83}Kr: ± 0.16%
^{84}Kr: ± 0.49%
^{86}Kr: ± 0.10%

QMS Noble Gas Quantification: Xenon Stable Isotopes

Sample size: ~3 nanomoles (10^{-9}) pure Xe gas



Preliminary External Reproducibility (1σ; *n* = 3)

^{124}Xe: ± 0.02%
^{126}Xe: ± 0.02%
^{128}Xe: ± 0.04%
^{129}Xe: ± 0.02%
^{130}Xe: ± 0.02%
^{131}Xe: ± 0.02%
^{132}Xe: ± 0.06%
^{134}Xe: ± 0.03%
^{136}Xe: ± 0.01%

Part 2

Daniel Eldridge

Crush Tests:
Demonstrating New Capability Development

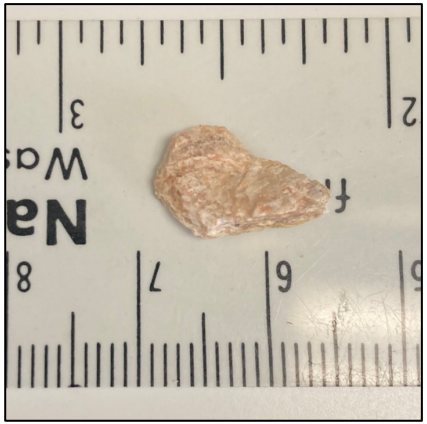
Crush Test 1:
Analog materials (Soda Dam Carbonate, ~300mg)
- Quantified gas composition by QMS (e.g., CO_2/N_2 ratio)
- Determined Carbon-13 isotope composition of liberated CO_2 by IRMS

Crush Test 2:
Experimental silicate melt glass containing two-phase inclusions (gas, aqueous fluid)
- Detected organic volatiles (contaminant) that inform ongoing experiments

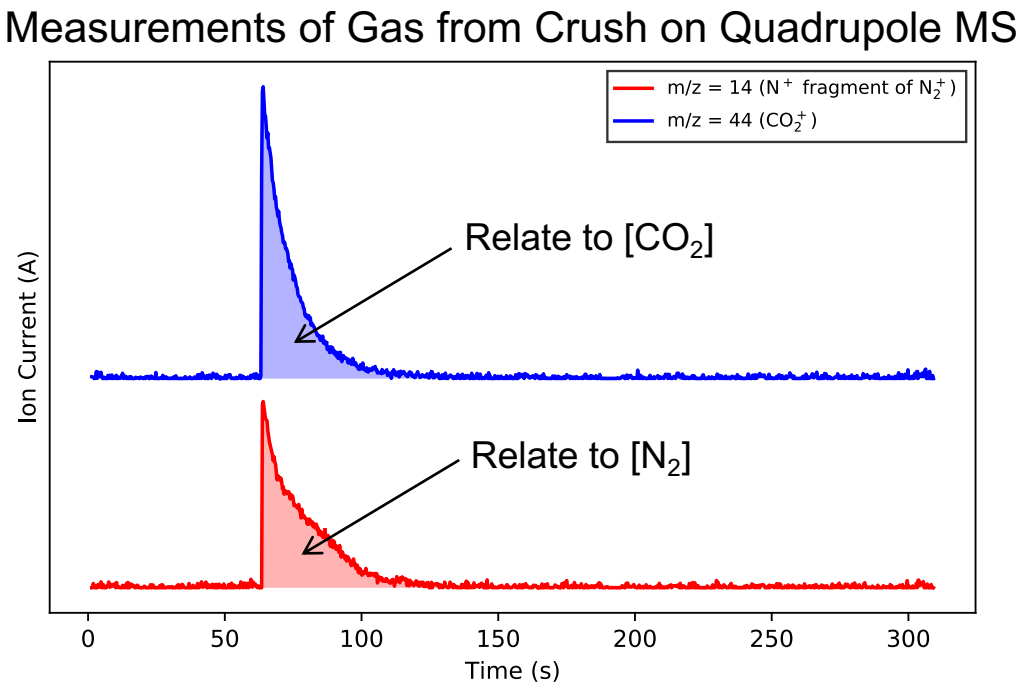
Crush Test 3:
Trinitite aerobead (~100mg)
- Detected gas signature unique to formation (fireball) environment

Crush Test 1: Local Soda Dam Carbonate (304 mg)

Background-corrected peaks at given m/z (e.g., N_2 and CO_2 quantification)

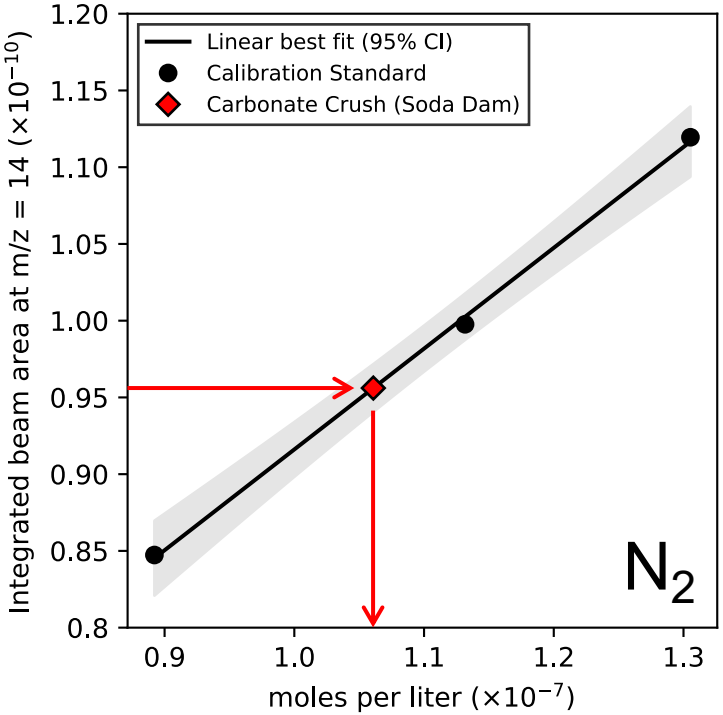


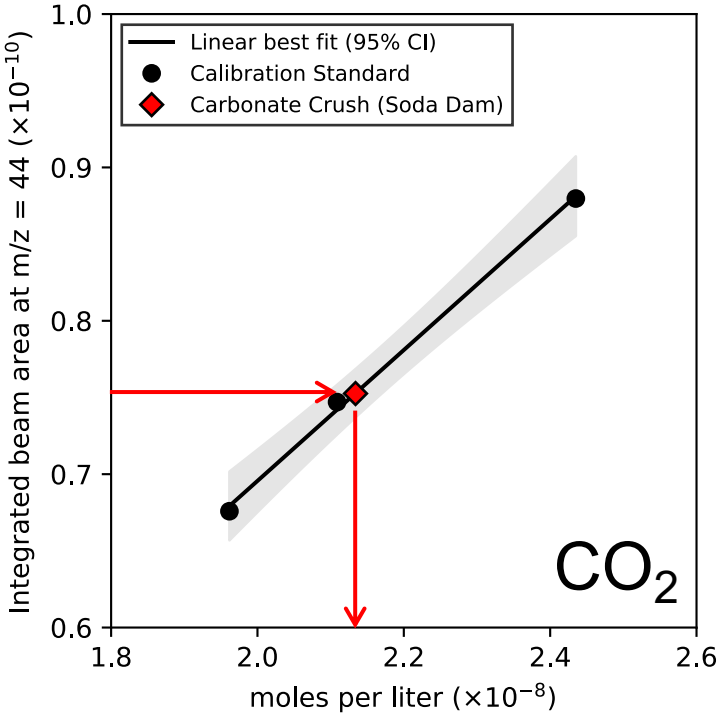
Soda Dam Carbonate Fragment



Crush Test 1: Local Soda Dam Carbonate (304 mg)

Localized 3-Point Calibration Based on Standard Gas

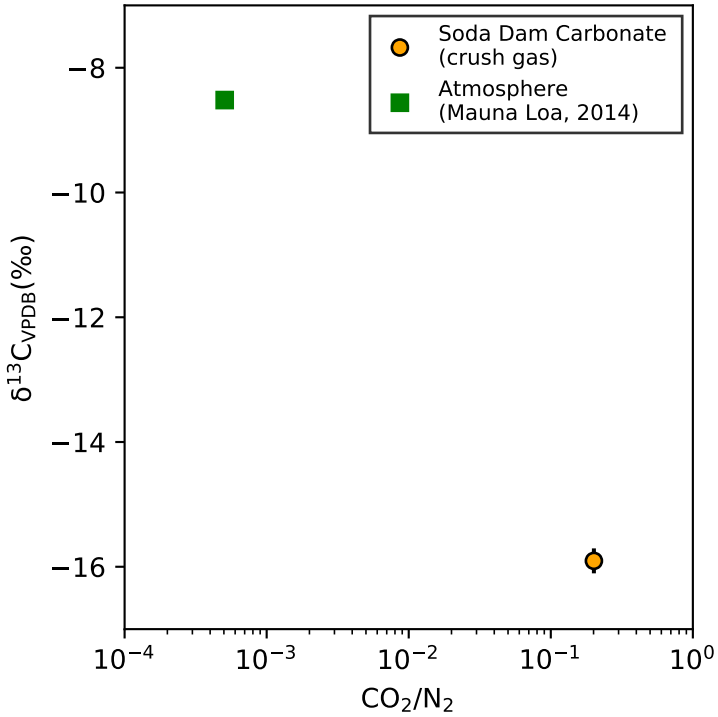




$$\frac{CO_2}{N_2} = 0.20 \pm 0.02 \text{ (1 s.e.)} \approx 400 \times Air$$

Crush Test 1: Local Soda Dam Carbonate (304 mg)

Stable Isotope Measurement (Carbon-13) of Crush-Liberated CO_2(g)

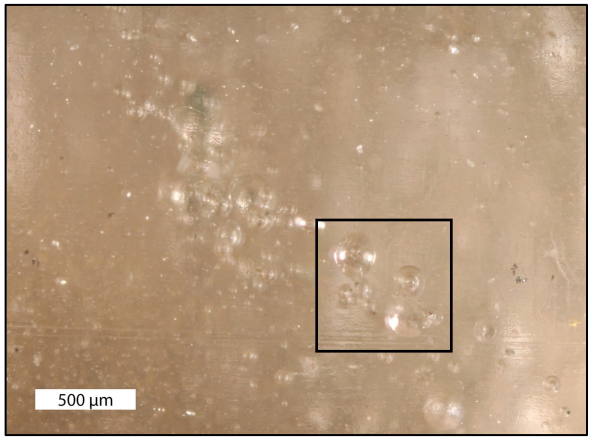


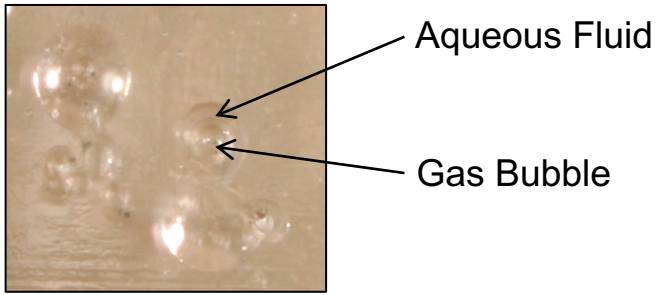
Atmospheric (NOAA-ESRL-GMD): http://www.esrl.noaa.gov/gmd

Crush Test 2: Experimental Silicate Melt

$NaAlSi_3O_8$-$KAlSi_3O_8$-SiO_2 System (haplogranite), H_2O-saturated, ~700˚C

Successfully generated melt glass with two-phase inclusions (aqueous fluid + gas)

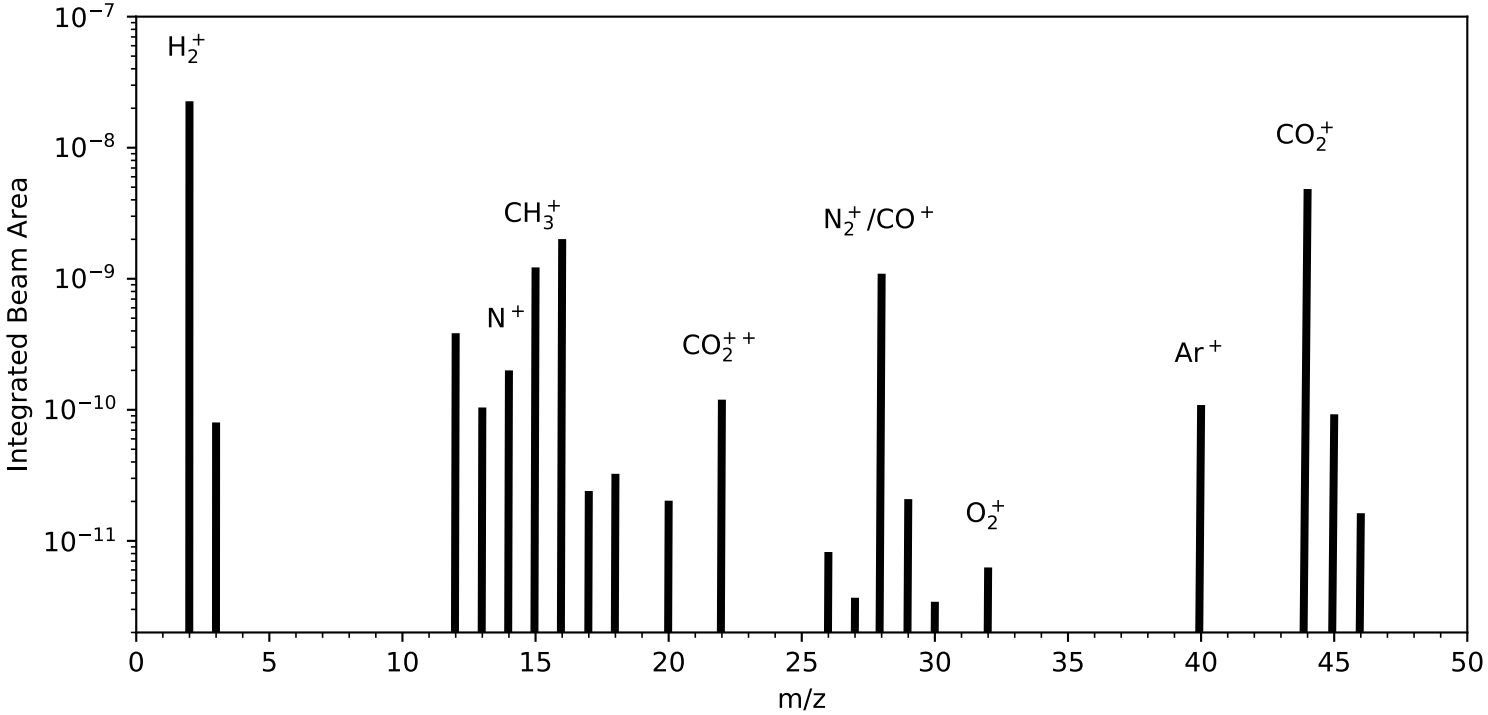




Experiments conducted by DR team-members:
Chris Campe (post-Masters student), Artaches Migdissov, Hakim Boukhalfa

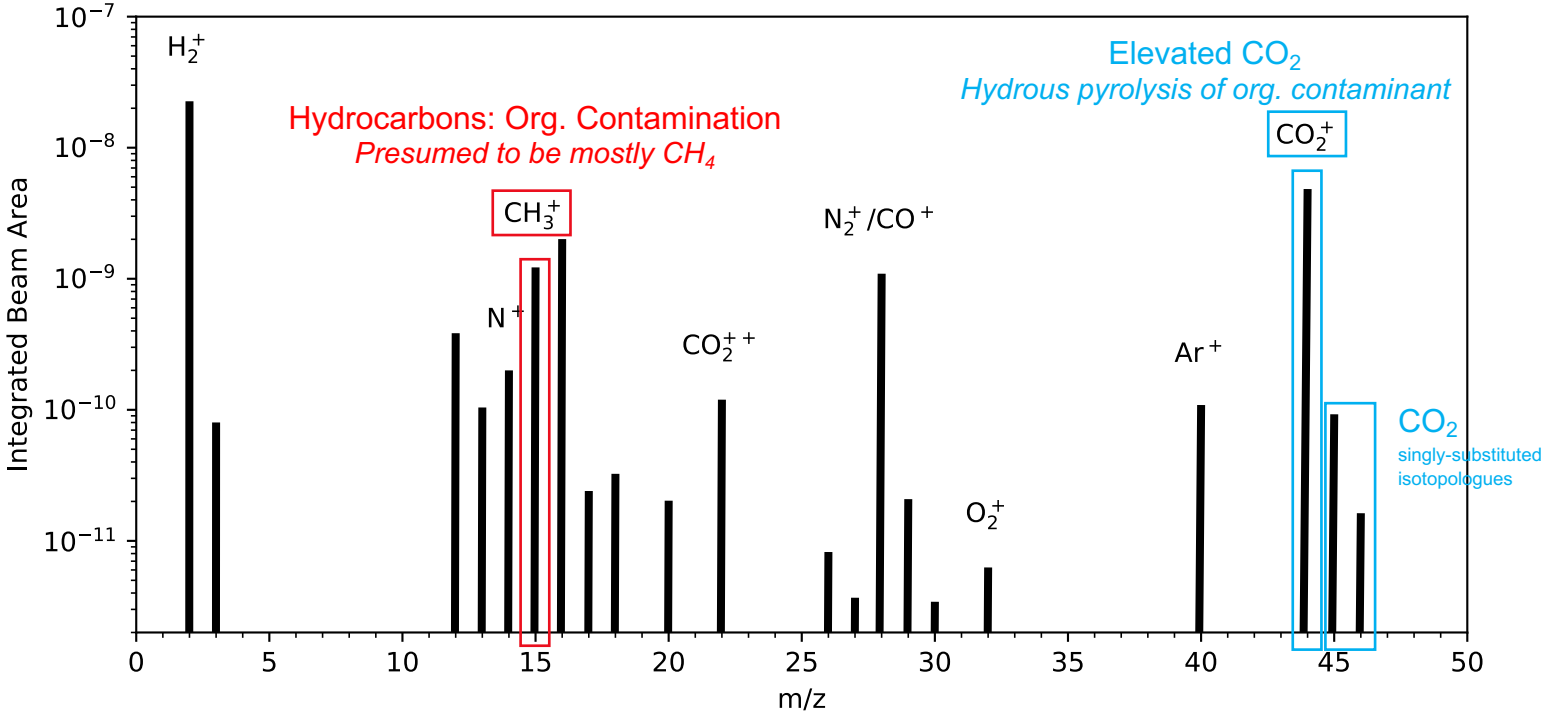
Crush Test 2: Experimental Silicate Melt

Mass spectrum of liberated gases from crush

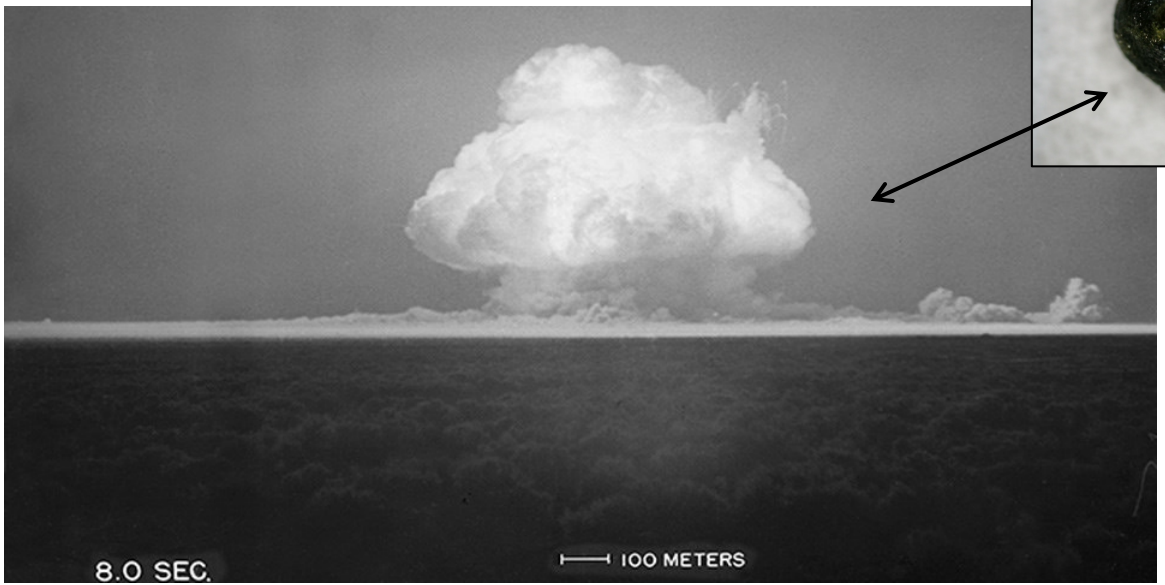


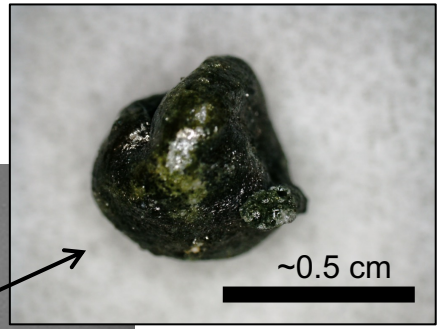
Crush Test 2: Experimental Silicate Melt

Identified organic contaminant in experiment (informs mitigation strategies in ongoing/future experiments)



Crush Test 3: Trinitite Aerobead (~100 mg)





National Park Service
https://www.nps.gov/whsa/learn/historyculture/trinity-site.htm

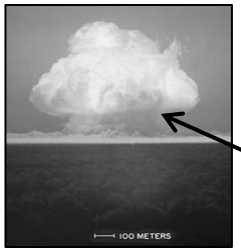
Crush Test 3: Trinitite Aerobead (~100 mg)

Crush Test 3: Trinitite Aerobead (~100 mg)

Detecting a CO(g)-rich formation environment?

Elevated H_2(g)*
Reducing Environment

C and O atomic fragments detected
but no CO_2(g) and little O_2(g):
Points to significant CO(g) in aerobead!*



***Sample of the reducing, H_2(g)- and CO(g)-rich fireball captured in aerobead (*new supporting observation of fireball conditions*)**

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Summary/Future

Accomplishments: Capability Development COMPLETE

- Developed and demonstrated capability for crushing materials and liberating volatiles (including trinitite).
- Demonstrated the ability to quantify (QMS) and perform stable isotope measurements (IRMS) on liberated gases.

Next steps: Measure Samples (~6 months) and Write-Up (~2 months)

- Expand measurements to more trinitite samples for open literature publication:
  - Aerobead *vs.* on-the-ground samples
  - Gas quantification + volatile metal isotope analysis on crushed material (e.g., Cs, Zn)
- Apply methodology to underground test materials.